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## Spatial and temporal variability in outdoor, indoor, and personal PM<sub>2.5</sub> exposure

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### Abstract

Outdoor, indoor and personal PM<sub>2.5</sub> measurements were made in a population of nonsmoking adults from three communities in the Minneapolis–St. Paul metropolitan area between April and November 1999. Thirty-two healthy adult subjects (23 females, 9 males; mean age  $42 \pm 10$ , range: 24–64 yr) were monitored for 2–15 days during the spring, summer, and fall monitoring seasons. Twenty-four hour average gravimetric PM<sub>2.5</sub> samples were collected using a federal reference monitor (Anderson RAAS2.5-300) located at outdoor (O) central sites in the Battle Creek (BCK), East St. Paul (ESP) and Phillips (PHI) communities. Concurrent 24-h average indoor (I) and personal (P), and a limited number of outdoor-at-home (O@H) samples were collected using inertial impactors (PEM™ Model 200, MSP, Inc). The O (geometric mean {GM} = 8.6;  $n = 271$ ; range: 1.0–41  $\mu\text{g}/\text{m}^3$ ) were lower than I concentrations (GM = 10.7;  $n = 294$ ; range 1.3–131  $\mu\text{g}/\text{m}^3$ ), which were lower than P concentrations (GM = 19.0;  $n = 332$ ; range 2.2–298  $\mu\text{g}/\text{m}^3$ ). Correlation coefficients between O concentrations in the three communities were high and measured GM O levels in BCK were significantly lower than ESP, most likely because of local sources, but GM concentrations in PHI were not significantly different from BCK or ESP. On days with paired samples ( $n = 29$ ), O concentrations were significantly lower (mean difference 2.9  $\mu\text{g}/\text{m}^3$ ;  $p = 0.026$ ) than O@H measurements (GM = 11.3; range: 3.5–33.8  $\mu\text{g}/\text{m}^3$ ), likely due to local sources in communities. Observed I and P concentrations were more variable, probably because of residential central air conditioning and hours of household ventilation for I and P, and occupational and environmental tobacco smoke exposures outside the residence for P. Across all individuals and days the median PM<sub>2.5</sub> “personal cloud” was 5.7  $\mu\text{g}/\text{m}^3$ , but the mean of the average for each participant was 15.7  $\mu\text{g}/\text{m}^3$ , with very low values in participants who did not work outside the home and much higher values in subjects with active lifestyles. Across all households and individuals the correlation between P and O concentrations was not significant, but the overall I–O correlation (0.27) and P–I correlation (0.51) were significant ( $p < 0.05$ ). Relatively little spatial variability was observed in O PM<sub>2.5</sub> concentrations across the three communities compared to the variability associated with I and P samples, and the measured O levels were relatively low compared to other large metropolitan areas in the United States. © 2002 Elsevier Science Ltd. All rights reserved.

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### 1. Introduction

Time-series epidemiological studies have shown a statistical association between day-to-day variability in outdoor particulate matter (PM) concentrations and

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mortality and morbidity (USEPA, 1996; Vedal, 1997; NRC, 1998; Samet et al., 2000). Most of these studies examined statistical relations between health outcomes and outdoor PM concentrations with mass median diameter of  $<10\mu\text{m}$  ( $\text{PM}_{10}$ ) measured at central monitoring sites in urban areas. While most of these epidemiological studies have been performed in urban areas with relatively high PM concentrations, studies have also indicated that a statistical relation exists between  $\text{PM}_{10}$  (and  $\text{PM}_{10}$  in concert with other pollutants) and hospital admissions for chronic lung diseases for the elderly in the Minneapolis–St. Paul Metropolitan area (Schwartz, 1994; Moolgavkar et al., 1997), which has relatively low ambient PM concentrations compared to other major urban areas in the United States (USEPA, 2001).

Critics of these epidemiological studies have noted that personal exposure monitoring studies have found only weak statistical associations between outdoor PM concentrations and personal exposure to PM because: (a) personal exposure to  $\text{PM}_{10}$  is typically greater than indoor concentrations (people typically spend more than 90% of their time indoors); (b) both personal and indoor  $\text{PM}_{10}$  concentrations are typically greater than outdoor concentrations; and (c) cross-sectional correlation coefficients (i.e., each day treated as an independent measurement) between outdoor concentrations and personal exposure are low (Ozkaynak et al., 1996a, b; Wallace, 1996).

More recent studies indicate that smaller particles, such as  $\text{PM}_{2.5}$  (mass median diameter  $<2.5\mu\text{m}$ ) may be more closely linked with health effects (Schwartz et al., 1996). In the United States the USEPA ambient  $\text{PM}_{2.5}$  monitoring network was established in 1999, but there are relatively few personal  $\text{PM}_{2.5}$  monitoring studies and most focus on subjects presumed to be more sensitive to PM health effects (Janssen et al., 1998, 2000; Ebelt et al., 2000; Evans et al., 2000; Rojas-Bracho et al., 2000; Sarnat et al., 2000; Williams et al., 2000a). Relatively few studies monitored healthy adults (Ozkaynak et al., 1996a, b; Brauer et al., 2000) and most studies have used a “panel” study design (i.e., participants monitored on consecutive days over a few weeks), as opposed to measurements within participants conducted over several seasons.

The objective of this study was to document outdoor, indoor, and personal  $\text{PM}_{2.5}$  levels in a population of healthy nonsmoking adults over multiple days and seasons for three communities in the Minneapolis–St. Paul metropolitan area. This paper describes the study design, data collection methods, population characteristics and the distribution of measured  $\text{PM}_{2.5}$  concentrations. It also examines the spatial and temporal variability within and between outdoor samples, factors associated with variability in indoor and personal samples, and the implications of these

results for assessing PM exposures in the general population.

## 2. Methods

### 2.1. Study design

This  $\text{PM}_{2.5}$  exposure study was conducted as part of research examining the statistical associations between a suite of hazardous air pollutants (volatile organic compounds (VOCs)) modeled for the Minneapolis–St. Paul metropolitan area and measured at outdoor central sites, within homes, and for adult subjects. Based on the results of ambient VOC modeling, the Battle Creek (BCK), East St. Paul (ESP), and Phillips (PHI) communities were chosen for monitoring: ESP and PHI were estimated to have relatively high and BCK relatively low VOC concentrations (Fig. 1) (Pratt et al., 1998). The selected communities varied in size and population density: PHI is the smallest ( $2.8\text{ km}^2$ ) but has the highest population density ( $2000\text{--}8000/\text{km}^2$ ), ESP is the largest area ( $18.2\text{ km}^2$ ) but less densely populated ( $1000\text{--}4000/\text{km}^2$ ) than PHI, and BCK is more than three times larger than PHI ( $9.8\text{ km}^2$ ), but with the lowest population density ( $500\text{--}2000/\text{km}^2$ ).

Healthy, nonsmoking adults were recruited within neighborhoods by house-to-house canvassing and direct solicitation. Each neighborhood was divided into equal-sized quadrants and participants were added to each quadrant sequentially to ensure a geographically diverse distribution of subjects around the outdoor central monitoring site. The greatest distance from a participating household to the central monitoring site in each community was 4.4 km in BCK, 3.5 km in ESP, and 1.1 km in PHI. The greatest distance between any two residences in a community was 4.9 km in both ESP and BCK, and 1.7 km in PHI. After informed consent was obtained, subjects completed a baseline questionnaire to determine smoking status, socio-demographics, occupation, housing characteristics, and a brief health history. After the first measurement session subjects were explicitly asked if they were able to continue carrying the PM monitoring equipment for the remainder of the study.

Monitoring sessions were conducted during the spring (26 April–20 June), summer (21 June–11 August), and fall (23 September–21 November) of 1999. Each monitoring session consisted of two consecutive 24-h periods, followed by a day to change filters, so that two sequential 24-h average outdoor central site (O), indoor (I), and personal (P)  $\text{PM}_{2.5}$  concentrations were obtained and a new sampling session was started every third day. Central site O samples were collected by technicians from the Minnesota Pollution Control Agency (MPCA) and samples were collected using

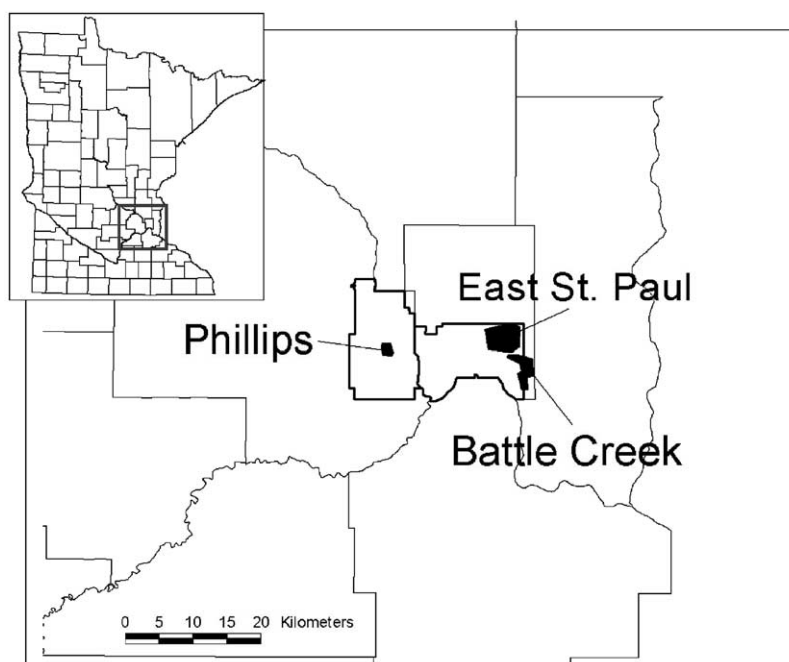


Fig. 1. Map of Minneapolis–St. Paul metropolitan area and location of the Battle Creek, East St. Paul, and Phillips communities.

monitors located near the approximate geographic center of each neighborhood using a federal reference monitor (FRM), which ran from 12:01 a.m. to 12 p.m. each day. Data collected in this study were part of the monitoring network maintained by MPCA as part of EPA's national  $\text{PM}_{2.5}$  monitoring network, the aerometric information retrieval system (AIRS). AIRS monitoring is typically conducted every third day, but 24-h average samples were collected for 2 days in a row (followed by an off day) to increase the number of monitored days for each participant.

A monitoring session consisted of two consecutive 24-h average samples. The P and I monitors were briefly stopped after the first 24-h sampling period to change filters and batteries (P monitors only), and re-grease impactor inlets. Monitoring sessions were conducted so that the two 24-h average I and P measurements were obtained concurrently with O samples for multiple individuals within each community on each day. Outdoor-at-home (O@H) measurements were also collected at randomly selected study households by placing a monitor on the front porch of participant home where P and I sampling was occurring. Up to 15 days of monitoring were collected per person, with a goal of at least two sampling sessions (4 monitored days) each season. For subject convenience and logistical reasons I, P, and O@H monitors were distributed and collected from subject homes in the evening (usually between 5 and 9 p.m.). Start times for P, I and O@H monitors were always within a few minutes of each other, so

comparisons between these measurements have essentially complete temporal overlap. For comparisons between the P, I, and O@H monitors with O monitors the average percent overlap (% overlap =  $1 - \{\text{Minutes P, I, or O@H sampler started before the O sampler start time} / 1440 \text{ min/day}\}$ ) were calculated for each set of measurement pairs.

Inlets for I monitors were placed in each subject's residence at approximately the subject's seated breathing height in the room where he/she reported spending the majority of their waking hours. Sampling pumps were kept in foam-insulated boxes. Subjects carried the personal samplers in small foam-insulated bags with a shoulder strap that had the inlet mounted on the front. During sampling sessions subjects were asked to wear or carry the exposure monitors whenever possible. Subjects were asked to place the monitor beside them while seated and to take it with them as they went about their activities. At night they were instructed to place it beside their bed.

On sampling days, subjects completed a time-activity diary (TAD), recording time spent in seven primary microenvironments (inside at home, work and/or school, and other; outside at home, work and/or school, and other; and in-transit). They also recorded data on exposure to tobacco smoke and other potential modifiers of exposure levels, such as occupational exposures, hobbies and the number of hours that doors and windows were open in a residence. Subject compliance with study protocols, such as completion of the TAD and carrying the monitor in accordance with

the study protocol appeared to be high based on random spot checks and recorded technician observations.

## 2.2. Sampling equipment and filter weighing

Gravimetric O concentrations were obtained using a FRM, the Anderson RAAS2.5-300 sampler (USEPA, 1997) and EPA site requirements for PM<sub>2.5</sub> samplers. Samples were collected on 46.2 mm, 2 µm pore size PTFE filters at a nominal flow rate of 16.7 l/min, and final concentrations were adjusted for passive loading (Ramachandran et al., 2000).

Gravimetric P, I and O@H concentrations were collected using PM<sub>2.5</sub> inertial impactor environmental monitoring inlets (PEM™ Model 200, MSP, Inc., Minneapolis, MN) (Marple et al., 1987) and air sampling pumps (Buck Genie Extra, A.P. Buck, Inc., Orlando, FL). Samples were collected on 37 mm PTFE, 2.0 µm pore size filters with a polyolefin support ring (Gelman Sciences, Ann Arbor, MI). Flow rates for the 24 h P and I/O@H samples were 4 and 10 l/min, respectively, and pump times (median 23:48 h; range 20:01–24:00 h) were used to calculate sample volumes. Pumps were calibrated before and after sampling using a primary gas flow standard (mini-Buck Calibrator Model M30, A.P. Buck Inc., Orlando, FL), and flows were checked at intermediate points. Samples with pre- or post-sampling flow rates that varied more than ±10% from target flow rates were deemed invalid and excluded from the final dataset.

All PM<sub>2.5</sub> filters were pre- and post-weighed according to EPA protocols on a microbalance (Cahn Instruments, Cerritos, CA) accurate to 1 µg in an environmentally controlled weigh room in accordance with the FRM (relative humidity between 30 and 40%, with a variability of not more than 5% over 24 h, and temperature between 20°C and 23°C, with a variability of not more than 2°C over 24 h). Filters weights were equilibrated in the controlled environment for at least 24 h prior to weighing. All filter weights were recorded twice: (1) after a 30 s stabilization time on the balance; and (2) after the initial weight was recorded, the balance was tared and the filter was removed, then the absolute value of this second weight was recorded. Ten percent of the filters were reweighed within 1 week, and two laboratory blank filters were weighed during each weighing session to monitor change in scale accuracy over time. Filters with holes or punctures from sampling or mishandling were deemed invalid and excluded from the final dataset.

## 2.3. Quality control and assurance

Approximately 10% of P, I and O@H filters were field blanks, and 16 field blanks were available from the FRM monitors. Field blanks were loaded into the

sampler inlet in an identical manner as the sample filter, but then removed immediately and stored at the sampling site for 24 h before returning to the lab with the sample filters. Mean changes in field blank weights were 10.9 µg ( $n = 16$ ; S.D. = 6.4) for O, 6.2 µg ( $n = 37$ , S.D. = 17 µg) for I and O@H and 7.1 µg ( $n = 42$ , S.D. = 14 µg) for P samples. The detection limit, defined as three times the standard deviation of the field blanks divided by the average sampled air volume, was 0.8 µg/m<sup>3</sup> for O, 3.6 µg/m<sup>3</sup> for I and O@H, and 7.5 µg/m<sup>3</sup> for P measurements. Mean field blank weights were subtracted from all sample weights prior to calculation of concentrations. One hundred percent of O, 95% of I, 97% of O@H and 90% of P concentrations were greater than their respective detection limits. Limited data exist on comparability between co-located sampler types, but the PEM inlets used for P and I sampling have been shown to be reasonably precise (USEPA, 1996). Research comparing samplers concluded that the PEM sampler displayed “a positive mass concentration bias ranging up to 18% relative to the FRM” (Williams et al., 2000b).

## 2.4. Statistical analysis

SAS® (Version 8.01, SAS Institute, Inc., Cary, NC) was used for tabulations and statistical analyses. Summary statistics were calculated by pooling all samples by type (O, O@H, I and P), then examining them by community and season. Normality of the underlying distributions was assessed using the Shapiro–Wilk statistic. Concentrations less than the detection limit were included in calculations of summary statistics (as opposed to substituting them with an arbitrary value). Because the distributions of all types of PM<sub>2.5</sub> concentrations were approximately log normally distributed, statistical comparisons between measures of central tendency were performed using log transformed values. Mean differences within and between communities and seasons were assessed using general linear models procedure (PROC GLM) and the least significant difference (LSD) and Tukey’s “honest significant difference” (HSD) procedures, while paired comparisons were made using *t*-tests.

Estimates of the “personal cloud” (PC) were modeled using PM<sub>2.5</sub> concentrations measured indoors and outdoors and time activity patterns (Rodes et al., 1991). We used the method developed for PTEAM (Ozkaynak et al., 1996b), in which modeled personal exposure (PE<sub>mod</sub>) is estimated using the formula  $PE_{mod} = C_I F_I + C_O F_O$ , where  $C$  is concentration and  $F$  is fraction of time spent indoors or outdoors ( $F_I + F_O = 1$ ), and PC (in µg/m<sup>3</sup>) =  $P - PE_{mod}$ . This method assumes that all indoor PM<sub>2.5</sub> concentrations in locations where participants spend time are estimated by the I measurement in a subject’s residence, that concentrations during time in transit are estimated by

the fixed site outdoor monitor, and the measurement error associated with PC is randomly distributed.

### 3. Results

A total of 32 healthy nonsmoking adult participants (23 females, 9 males; mean age  $42 \pm 10$ , range: 24–64 yr) were monitored during the spring, summer, and fall monitoring seasons. Seven (6 females) of the 32 subjects reported that they did not work outside the home. The other 25 subjects had a wide variety of occupations, including several who worked in environments with known exposures to dust or fumes. All subjects were nonsmokers, and only one reported living with a smoker (who did not smoke inside their residence). Twenty-eight of the 32 subjects were monitored until the end of the study, with one dropping out after the spring season, and three dropping out after the summer season.

#### 3.1. Sample capture

A total of 271 O samples were obtained and at least one valid sample was obtained in at least one of the three

communities on 111 of 112 monitored days (Table 1). A total of 294 valid I, 38 valid O@H and 332 valid P samples were obtained, with the largest proportion of invalid samples occurring during the spring season due to pump malfunctions. No valid I or O@H samples were obtained between 4 and 14 June because of equipment failures, and pumps were subsequently modified to increase reliability. Thereafter the percentage of valid samples increased substantially. For example, the percentage of valid I samples increased from 62% to 86% to 90% in the spring, summer, and fall seasons, respectively. Similarly, the percentage of valid P samples increased from 73% to 81% to 88% in the spring, summer, and fall seasons, respectively.

#### 3.2. Central site outdoor concentrations

The distribution of O measurements was positively skewed and approximately log normal within each community. The geometric mean (GM) of all measurements was  $8.6 \mu\text{g}/\text{m}^3$ . Concentrations were highly correlated between communities (Fig. 2) and log scale bivariate Pearson correlation coefficients were all statistically significant ( $p < 0.001$ ): 0.85 (BCK versus

Table 1

Descriptive statistics for outdoor, indoor, and personal  $\text{PM}_{2.5}$  concentrations stratified by community and season (all values in  $\mu\text{g}/\text{m}^3$ , except as indicated)

	Battle Creek						E. St. Paul						Phillips					
	<i>N</i> <sup>a</sup>	Mean	S.D.	GM <sup>b</sup>	GSD <sup>c</sup>	Range	<i>N</i>	Mean	S.D.	GM	GSD	Range	<i>N</i>	Mean	S.D.	GM	GSD	Range
<i>Outdoor</i>																		
All Seasons <sup>d</sup>	88	9.4	6.2	7.8	1.8	1.0–35.4	95	10.8	6.6	9.3	1.8	1.1–41.6	88	10.0	5.8	8.7	1.7	2.8–21.6
Spring	36	10.5	7.1	8.5	2.0	1.0–33.1	36	12.0	7.3	10.1	1.9	1.1–35.5	30	12.1	7.2	10.5	1.7	3.6–35.3
Summer	22	8.7	4.4	7.8	1.6	3.5–20.0	25	8.5	3.2	7.8	1.6	2.3–14.4	30	8.6	3.8	7.8	1.6	2.8–16.9
Fall	30	8.4	6.2	7.1	1.7	2.1–35.4	34	11.3	7.5	9.6	1.8	2.5–41.6	28	9.3	5.5	8.1	1.7	2.8–27.9
<i>Indoor</i>																		
All Seasons <sup>e</sup>	108	10.6	6.6	9.0	1.8	2.3–36.9	97	17.4	20.3	12.2	2.2	1.3–130	89	14.2	13.0	11.3	1.9	3.1–90.9
Spring	25	12.7	7.7	11.0	1.7	4.7–35.5	30	20.7	26.4	13.6	2.4	3.0–130	15	16.9	14.2	13.0	2.1	4.9–60.8
Summer	36	8.9	3.8	8.1	1.5	3.5–16.4	26	15.8	11.4	13.7	1.6	4.7–65.9	36	13.2	6.4	11.4	1.7	3.1–30.2
Fall	47	10.9	7.4	8.8	2.0	2.3–36.9	41	16.0	19.6	10.4	2.4	1.3–97.7	38	14.4	16.7	10.6	2.0	3.3–90.9
<i>Personal</i>																		
All Seasons <sup>f</sup>	118	22.6	25.7	16.2	2.2	3.8–207	107	30.5	38.7	20.6	2.3	2.5–298	107	26.5	24.3	20.9	2.0	2.2–211
Spring	41	26.3	25.7	19.4	2.1	3.9–133	44	33.9	34.4	23.9	2.3	2.5–201	28	37.5	37.6	30.0	1.8	14.9–211
Summer	31	28.5	36.1	20.3	2.1	5.9–207	25	20.5	15.0	17.2	1.8	5.9–82.4	40	22.6	15.3	19.2	1.7	2.7–82.3
Fall	46	15.5	13.4	11.9	2.1	3.8–80.3	38	33.1	51.9	19.5	2.5	5.0–298	39	22.6	16.7	17.6	2.1	2.2–67.5

<sup>a</sup>Number of valid observations.

<sup>b</sup>Geometric Mean.

<sup>c</sup>Geometric Standard Deviation.

<sup>d</sup>336 total outdoor samples attempted, with 65 (19%) invalidated because of equipment failure.

<sup>e</sup>367 total indoor samples attempted, with 62 (16.9%) indoor of filters invalidated because of pump problems (e.g., flows outside of target range), and 19 (5.2%) of samples invalidated because of filter problems (e.g., punctures, mishandling).

<sup>f</sup>413 total personal samples were attempted, with 38 (9.2%) filters invalidated because of pump problems (e.g., flows outside of target range, battery problems) and 44 (11%) of personal filters invalidated because of filter problems (e.g., punctures, mishandling).

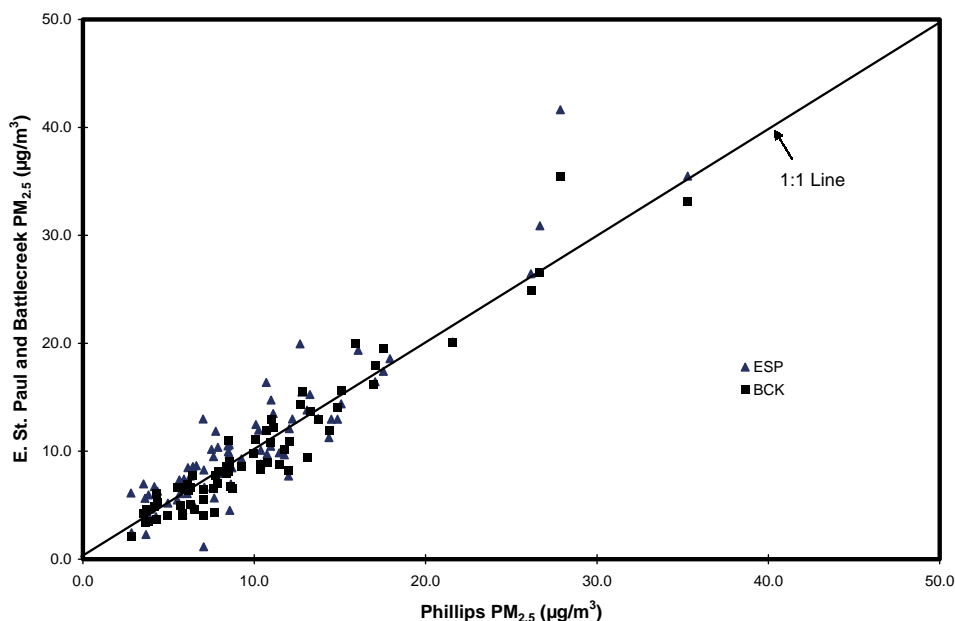


Fig. 2. Outdoor  $PM_{2.5}$  concentrations ( $\mu\text{g}/\text{m}^3$ ) measured at central sites in Battle Creek (BCK) and East St. Paul (ESP) plotted against Phillips (PHI).

ESP), 0.94 (BCK versus PHI), and 0.83 (ESP versus PHI). Geometric mean levels in BCK were significantly lower than GM levels observed in ESP (LSD procedure;  $p < 0.05$ ) (Table 1), although these GMs are not significantly different if the more conservative HSD procedure is used ( $p > 0.05$ ). There was no significant difference between GM levels in PHI and in the BCK or ESP communities. On days with paired samples, log concentrations in BCK were significantly lower than in ESP ( $n = 78$ ;  $t = 3.19$ ;  $p = 0.0021$ ), but BCK was not significantly different from PHI ( $n = 70$ ;  $t = 1.9$ ;  $p = 0.062$ ), and ESP was not significantly different from PHI ( $n = 78$ ;  $t = 1.66$ ;  $p = 0.10$ ). Geometric mean O concentrations were significantly higher in the spring compared to the summer ( $p < 0.05$ ), while spring–fall and summer–fall GMs were not significantly different ( $p > 0.05$ ). Adjusting for the effect of community did not change these results.

### 3.3. Outdoor-at home measurements

A total of 38 valid O@H samples were obtained over three seasons, and there were 29 days for which these could be paired with a valid O sample in the same community. On these paired days the GM of O@H samples was  $11.3 \mu\text{g}/\text{m}^3$  (range: 3.5–33.8), and O@H measurements were significantly higher than O measurements collected on the same day (O@H–O mean difference =  $2.9 \mu\text{g}/\text{m}^3$ ;  $t = 2.35$ ,  $p = 0.026$ ).

### 3.4. Indoor concentrations

The distribution of I measurements was positively skewed, with an overall GM of  $10.7 \mu\text{g}/\text{m}^3$ . We obtained a mean of 9 (range 2–13) I measurements per household, and 7 or more days of data in 28 of the 32 households monitored.  $PM_{2.5}$  concentrations inside the residence of the single participant who reported living with a smoker (who smoked outside the residence) were somewhat higher than average I values but not substantially elevated ( $n = 10$ ; mean =  $15.9$ ; range  $7.7$ – $21.7 \mu\text{g}/\text{m}^3$ ). Geometric mean I concentrations in BCK ( $9.0 \mu\text{g}/\text{m}^3$ ) were significantly ( $p < 0.05$ ) lower than levels observed in ESP ( $12.2 \mu\text{g}/\text{m}^3$ ) and PHI ( $11.3 \mu\text{g}/\text{m}^3$ ) (Table 1). The BCK and PHI GMs were not significantly different ( $p > 0.05$ ). If the more conservative HSD statistic was used, only the difference between BCK and ESP concentrations are significant ( $p < 0.05$ ).

There was an average of 3 (range: 0–7) I measurements per day for the 112 monitoring days. The GM of summer season I measurements in BCK was significantly lower ( $p < 0.05$ ) than the GM of summer I measurements in ESP and PHI. Ten of the 12 households (83%) in BCK had central air conditioning, while only 2 households in ESP (18%) and 2 in PHI (22%) had central air conditioning. As a consequence, households in ESP and PHI were much more likely to have their doors or windows open (Table 2), especially during the summer monitoring season. Geometric mean I concentrations were also significantly higher in the

Table 2

Summary of individual time-activity patterns, tobacco exposure, and household ventilation patterns. Results reported as hours per day unless otherwise indicated

Variable	Mean	S.D.	Median	Min	Max
<i>Time spent in microenvironment</i>					
Indoors (all locations)	21.6	2.4	22.0	10.0	24
at home	17.2	4.7	18.0	1.0	24
at work/school	3.1	4.3	0	0	15.4
at other	1.3	1.9	0	0	9.5
Outdoors (all locations)	2.8	3.6	1	0	15.4
at home	0.7	1.4	0	0	9.0
at work/school	1.6	3.4	0	0	15.4
at other	0.5	1.3	0	0	8.5
In transit	1.0	1.0	0.9	0	8.5
Minutes/day tobacco exposure <sup>a</sup>	14	62	0	0	600
Battle Creek	0.8	8.3	0	0	90
East St. Paul	38	102	0	0	600
Phillips	5.9	19	0	0	120
Hours of household ventilation <sup>b</sup> (all locations)	9.7	10.4	4.0	0	24
Battle Creek	6.9	8.9	2.0	0	24
East St. Paul	13.7	10.6	15.0	0	24
Phillips	9.9	10.4	4.0	0	24

<sup>a</sup> Measured on days with a valid personal PM<sub>2.5</sub> measurement.

<sup>b</sup> Hours per day that windows and or doors open.

spring compared to the fall ( $p < 0.05$ ), but spring–summer and summer–fall mean differences were not significantly different ( $p > 0.05$ ).

### 3.5. Personal concentrations

A total of 332 valid P samples were obtained from the 32 subjects (Table 1). The distribution of P concentrations was positively skewed, with an overall GM of  $19.0 \mu\text{g}/\text{m}^3$ . The mean number of days per person with a valid P sample was 10, and 29 of the 32 subjects had 7 or more days of measurements. Almost all participants with the highest measured concentrations recorded tobacco exposures during their daily activities outside the home (Table 2), and many recorded occupational exposures to dusts or fumes on their time-activity diaries. Using the population median values shown in Table 2, individuals spent more than 90% of their time indoors (and more than 70% of their time indoors at home). The remaining 10% of their time was split between outdoors or in transit.

Geometric mean P concentrations in BCK were significantly ( $p < 0.05$ ) lower than levels observed in ESP and PHI, and this pattern was consistent across all seasons (Table 1). Geometric mean concentrations for residents of ESP and PHI were not significantly different from each other, but subjects from these communities

reported more minutes of tobacco exposure (either at work, at non-home indoor locations, or outdoors) than subjects who lived in BCK. Seven ESP participants reported nonzero minutes of tobacco exposure on a total of 26 days, including one subject who recorded tobacco exposure on 12 days, with an average of 248 min of exposure per day. Seven PHI subjects reported nonzero minutes of tobacco exposure on 15 days, while only 3 BCK subjects reported nonzero tobacco exposure on 3 days. Reversing the pattern observed for the O and I samples, the GM of P exposures were lowest in the spring and highest in the fall.

### 3.6. Percent temporal overlap between measurements

While the P, I, and O@H measurements all had similar start times within a household on a monitored day, typically in the later afternoon, the matched O measurement always started the following midnight. The mean percent temporal overlap between matched P/I and O measurements was  $71 \pm 9.0\%$  for P ( $n = 332$ ) and  $72 \pm 8.3\%$  for I ( $n = 294$ ). If the 25 days with  $< 50\%$  overlap are excluded the values increase to  $72 \pm 7.1\%$  for P ( $n = 317$ ) and  $73 \pm 6.7\%$  for I ( $n = 284$ ). For all the P and/or I measurements with more than 50% overlap with the central site O measurement the median overlap was 72% ( $n = 601$ ; range 51–90%). The combined P and

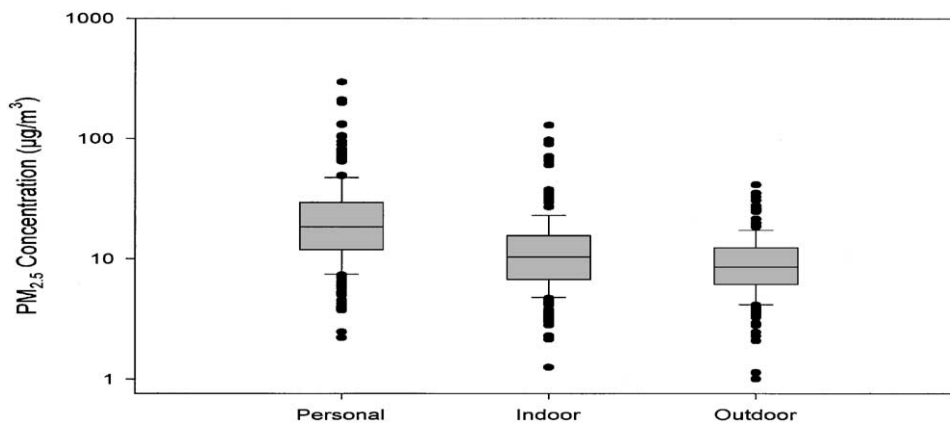


Fig. 3. Box plot of personal, indoor and outdoor  $PM_{2.5}$  concentrations ( $\mu\text{g}/\text{m}^3$ ) for all communities and seasons. Boxes indicate median, interquartile range, and dots indicate values outside that range.

I distribution of the overlaps was positively skewed, and the 10th and 90th percentiles of the distribution were 61% and 80%, respectively. Mean overlap between O and O@H samples was  $75 \pm 6\%$  ( $n = 29$ ; range 61–84%).

### 3.7. Personal cloud estimates

The PC was estimated for all days with valid matched P and I measurements ( $n = 239$ ). The pooled distribution of PCs for all subjects has a mean value of  $11.5 \mu\text{g}/\text{m}^3$  and a median of  $5.7 \mu\text{g}/\text{m}^3$  (range  $-64.5$ – $198.3 \mu\text{g}/\text{m}^3$ ); large positive values were observed for subjects who recorded occupational or ETS exposures outside the home on their time-activity diaries. The mean of the average PC for each subject was  $15.3 \mu\text{g}/\text{m}^3$  (range 0.7–67.8). PC levels were highest in participants who worked outside the home and had more active lifestyles, as indicated by the positive correlation coefficient between the percentage of time spent outdoors and the PC ( $r = 0.32$ ,  $p < 0.0001$ ). The top three mean PC values (all  $> 60 \mu\text{g}/\text{m}^3$ ) were in male participants who led active lifestyles, but these values represent a total of 6 monitored days. At the low end, 5 of the 6 lowest mean PC values were in female participants, 4 of whom did not work outside the home.

### 3.8. Statistical associations between outdoor, indoor, and personal concentrations

The overall distribution of P, I, and O samples are shown in Fig. 3. The relatively high correlation coefficients and small absolute differences between O concentrations among the 3 communities allows us to estimate missing values in a community using the mean value from the other two communities ( $n = 28$  days) or by the single community for which a valid sample was

available ( $n = 17$  days). Where necessary, this estimation procedure was used in the comparisons between O, I, and P concentrations. Across all communities and seasons average P concentrations were higher than both O and I concentrations, and average I concentrations were higher and more variable than average O concentrations.

The log correlation between matched I and O measurements (I–O) in all households on all days was relatively low (0.27) but statistically significant (Table 3). If this analysis is stratified by community the I–O correlation was higher in BCK and PHI compared to ESP, but the I–O correlation in ESP increases slightly if the subject with the most minutes of reported tobacco exposure is removed from the analysis. The I–O correlation coefficient was also higher for spring and summer compared to the fall season (Table 3). The log correlation between matched P and O measurements for all subjects was low and not statistically significant, even if the analysis was stratified by community or season. The log correlation between matched P and I (P–I) measurements was moderately high (0.51) and statistically significant (Table 3). The P–I log correlation was much higher in ESP compared to BCK and PHI, and P–I correlation was higher in the fall compared to the spring and summer seasons (Table 3).

## 4. Discussion

Our data are consistent with the general pattern observed in most PM monitoring studies: O concentrations are lower than I, which are lower than P exposures. Although O levels measured in this study are among the highest in the state of Minnesota, they are relatively low compared to other large urban areas in the United States. Ambient monitoring data from the USEPA



Table 3

Log correlations (*r*) between outdoor (O), indoor (I), and personal (P) PM<sub>2.5</sub> concentrations on matched days

	I–O		P–O		P–I	
	<i>r</i>	<i>p</i> =	<i>r</i>	<i>p</i> =	<i>r</i>	<i>p</i> =
All households	0.27	<0.0001	0.06	0.29	0.51	<0.0001
<i>By community (all seasons)</i>						
Battle Creek	0.40	<0.0001	0.02	0.85	0.37	0.0005
East St. Paul	0.13	0.22	0.06	0.55	0.72	<0.0001
Results with one high exposure subject removed <sup>a</sup>	0.19	0.042	0.20	0.06	0.63	<0.0001
Phillips	0.35	0.0008	0.13	0.20	0.40	0.0002
<i>By season (all communities)</i>						
Spring	0.34	0.005	0.14	0.14	0.40	0.004
Summer	0.32	0.001	0.08	0.46	0.44	<0.0001
Fall	0.20	0.03	0.03	0.76	0.60	<0.0001

<sup>a</sup>One East St. Paul participant had up to 600 min per day of tobacco exposure recorded on their time-activity diary.

indicate that in 1999 the Minneapolis–St. Paul Metropolitan Statistical Area (MSA) ranked 25th out of the 30 largest MSAs in the United States, with an annual average of 11.3 µg/m<sup>3</sup> (USEPA 2001). Annual average ambient concentrations in the Minneapolis–St. Paul MSA were less than half of the top two MSAs, Atlanta, GA (23.8 µg/m<sup>3</sup>) and Fresno, CA (23.0 µg/m<sup>3</sup>).

The log correlation between O concentrations in the three communities was higher than typically observed for PM<sub>10</sub>, presumably because of the variability introduced by the PM<sub>2.5–10</sub> fraction of the aerosol. The range of O PM<sub>2.5</sub> concentrations observed in this study is relatively small compared to other metropolitan areas, thus there is less overall variability to work with when calculating associations between ambient PM concentrations and measures of mortality or morbidity. Nevertheless, previous studies in the Minneapolis–St. Paul metropolitan area have observed associations between health effects and the PM<sub>10</sub> fraction of the aerosol (or PM<sub>10</sub> in concert with other pollutants) measured at central monitoring locations (Schwartz, 1994; Moolgavkar et al., 1997). In this study we had three O monitoring sites, and observed statistically significant differences in GM levels at the two closest measurement sites (BCK and ESP), but not between PHI and BCK or PHI and ESP. These observed differences are likely due to local sources and minor differences in meteorology between communities. This contention is supported by the observation that within-day 15-min average outdoor concentrations during this study varied by as much as an order of magnitude and were strongly influenced by changes in wind direction (Ramachandran et al., 2002). The effect of local sources is also seen on days with paired O and O@H samples: O concentrations were significantly lower than O@H measurements.

The I concentrations observed in this study were higher than O concentrations, and the observed levels appear to be similar to other nonsmoking residences in North America (Pellizzari et al., 1999; Rojas-Bracho et al., 2000). The between-community and season variability in observed I concentrations in this study are probably due to the presence of central air conditioning, differences in the hours of ventilation within households, and particle generating activities by residents. Similarly, P levels did not vary significantly across seasons but did vary by community, with much of the variability apparently driven by time spent in the presence of smokers and occupational exposures. While median PM<sub>2.5</sub> PC levels are higher than levels observed in recent studies of compromised adults (Williams et al., 2000a), both the upper and lower extremes of the PC distribution are clearly affected by human activities, and the upper extreme may also be affected by the contribution of unmeasured indoor exposures outside the home.

There are two facets of our study design that limit our ability to generalize these results to other PM exposure studies. First, our participants were a convenience sample of healthy nonsmoking adults from three different communities monitored over three seasons. The PM sampling in this study was thus conducted over a longer time period than most exposure panel studies conducted to date, in a more heterogeneous population with a wider variety of ages and occupations, and in a population with more than twice as many female as male subjects. Second, it is also important to note that the 24-h average P and I samples, which typically started between 5 and 7 p.m., did not have complete temporal overlap with the 24-h average O samples, which all started at midnight. The average overlap between P/I and O samples was 72%, and this temporal

discontinuity introduces an additional level of uncertainty into associations between measurements. While it is not feasible to quantify this uncertainty, the inherent measurement error of the P and I gravimetric PM<sub>2.5</sub> samplers is substantial. If expressed as the ratio of standard deviation of the field blanks over the average sampling volume (=MDL/3) divided by the GM and converted to % for each sampler type (i.e.,  $P = (2.5/19) = 13\%$ ;  $I = (1.2/10.7) = 11\%$ ;  $O = (0.27/8.6) = 3.1\%$ ), the measurement error for P and I samples is about half the magnitude of the error introduced by the temporal offset. The actual error introduced by the temporal offset may be less than it appears because of the moderately high correlation coefficients between O PM<sub>2.5</sub> concentrations in each community and the next day's O measurement (i.e., autocorrelation). These correlation coefficients were 0.45 ( $n = 45$ ;  $p = 0.002$ ) for PHI, 0.46 ( $n = 47$ ;  $p = 0.001$ ) for ESP, and 0.52 ( $n = 45$ ,  $p = 0.0002$ ) for BCK.

The overall correlation coefficients obtained by treating each measurement as independent were relatively high for P and I, moderate for I and O, and low and not statistically significant for P and O. The relatively high correlation coefficients between P and I concentrations probably occur because of the combination of complete temporal overlap and because study participants spent a large proportion of time indoors at home, a result consistent with population-based time activity studies and other PM monitoring studies. Although PC estimates varied substantially in the overall population, the estimate of average PM<sub>2.5</sub> PC measurements in individual participants was larger than the population median, and substantially higher in participants who spent more time outdoors or who had more active lifestyles. The moderate I–O correlation is likely the result of the temporal offset between samples and indoor source modification, which is consistent with the fact that correlation coefficients were higher during the spring and summer months when windows and doors were more often open. This assertion is also supported by the observation that I–O correlation coefficients were lowest during the fall, which had the lowest number of hours with windows and doors open. The relatively low P–O correlation is likely the result of P activities and occupational exposures that vary substantially both between- and within-subjects. Understanding within-person variability over time is an area where our understanding is expanding (Ebel et al., 2000; Janssen et al., 2000; Rojas-Bracho et al., 2000).

## 5. Conclusions

The O PM<sub>2.5</sub> concentrations observed in this study were all below the USEPA's PM<sub>2.5</sub> 24-h average

standard ( $65 \mu\text{g}/\text{m}^3$ ) and were relatively low compared to other major cities in the United States. Based on our analysis of three spatially separated monitors, a centrally located ambient PM<sub>2.5</sub> monitor will likely estimate the annual O average for the Minneapolis–St. Paul metropolitan area with reasonable accuracy. These I and P PM<sub>2.5</sub> measurements provide an estimate of the mean and variability for I and P concentrations for a population of healthy nonsmoking adults. The range of and variability in O concentrations in this study was low compared to I concentrations, which were in turn lower than the range and variability in P concentrations in this nonsmoking population. The statistical association between P and I concentrations were relatively strong, while I and O concentrations were moderately correlated across individuals. There was no observed statistical relation between P and O concentrations most likely because of the high between person variability and relatively low variability in O concentrations.

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